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Strong Correlations and Orbital Texture in Single-Layer 1T-$TaSe_2$

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Abstract:

Strong electron correlation can induce Mott insulating behavior and produce intriguing states of matter such as unconventional superconductivity and quantum spin liquids. Recent advances in van der Waals material synthesis enable the exploration of Mott systems in the two-dimensional limit. Here we report characterization of the local electronic properties of single- and few-layer 1T-$TaSe_2$ via spatial- and momentum-resolved spectroscopy involving scanning tunneling microscopy and angle-resolved photoemission. Our results indicate that electron correlation induces a robust Mott insulator state in single-layer 1T-$TaSe_2$ that is accompanied by unusual orbital texture. Interlayer coupling weakens the insulating phase, as shown by reduction of the energy gap and quenching of the correlation-driven orbital texture in bilayer and trilayer 1T-$TaSe_2$. This establishes single-layer 1T-$TaSe_2$ as a useful platform for investigating strong correlation physics in two dimensions.

Two-dimensional (2D) Mott insulators emerge when the Coulomb interaction (U) exceeds the bandwidth (W) in partially-filled band systems that can be described by 2D Hubbard-like models[1]. Correlated electronic behavior in quasi-2D Mott insulators leads to collective quantum phenomena[2,3] such as high-temperature superconductivity which is widely believed to arise through doping of Mott insulating copper-oxygen layers[4,5]. Certain stacked graphene systems have also recently been found to exhibit Mott-like insulating behavior and unconventional superconductivity upon gating[6-9]. Layered transition metal dichalcogenides (TMDs) offer another family of correlated quasi-2D materials, two examples being bulk 1T-TaS_2 and the surface of bulk 1T-$TaSe_2$ which have long been known to host unusual insulating phases in the star-of-David charge density wave (CDW) state[10-13]. Although widely believed to be Mott insulators[11,14,15], the insulating nature of these bulk systems is complicated by interlayer CDW stacking whose effects on the insulating phase remain controversial[16]. Interlayer hopping (which increases W) and interlayer dielectric screening (which decreases U) are expected to suppress Mott insulating behavior[1,17,18], but orbital stacking has also been predicted to open a hybridization gap even in the absence of electron correlation[16,19,20].

Atomically-thin 1T-TMDs offer an ideal platform to differentiate the contributions of electron correlation and interlayer effects in quasi-2D materials since single-layer systems can be fully characterized in the absence of interlayer coupling. Without interlayer coupling the reduced screening environment of a single-layer leads to increased Coulomb interaction and potentially enhanced correlation phenomena[21-25]. The effects of interlayer coupling can then be systematically determined by adding new layers to a material one at a time and mapping out the resulting stacking order and wavefunction properties. Previous studies on single-layer 1T-$NbSe_2$ and 1T-$TaSe_2$ have found unusual insulating behavior[26,27], but electronic wavefunction texture

and interlayer coupling effects were not examined. The nature of the insulating phase in these single-layer materials thus remains inconclusive.

Here we report a combined scanning tunneling microscopy/spectroscopy (STM/STS), angle-resolved photoemission spectroscopy (ARPES), and theoretical study of the electronic structure of single-layer 1T-$TaSe_2$. Our results show that in the absence of interlayer coupling single-layer 1T-$TaSe_2$ hosts a Mott-insulating ground state that exhibits a 109 ± 18 meV energy gap and unusual orbital texture. Bilayer and trilayer 1T-$TaSe_2$ with shifted stacking order exhibit successively smaller energy gaps and show no signs of the unusual orbital texture seen in the single-layer limit. The single-layer band structure and density of states of 1T-$TaSe_2$ are found to be consistent with DFT+U calculations, confirming its Mott insulator nature. The unusual single-layer orbital texture, however, is not captured by DFT+U, but is consistent with the behavior expected for a weakly screened, strongly correlated 2D insulator. Reduction of the 1T-$TaSe_2$ bandgap and quenching of the unusual orbital texture by the addition of new layers shows that the effect of interlayer coupling on shifted-stacked 1T-$TaSe_2$ is to weaken the Mott behavior. The single-layer limit of 1T-$TaSe_2$ is thus unique in that strong correlation effects here are most pronounced, affecting both the energy gap and electron wavefunction symmetry.

Electronic structure of single-layer 1T-$TaSe_2$ in the CDW phase

Our experiments were carried out on 1T-$TaSe_2$ thin films grown by molecular beam epitaxy on epitaxial bilayer-graphene-terminated (BLG) 6H-SiC(0001), as sketched in Fig. 1a. The crystal structure of 1T-$TaSe_2$ consists of a layer of Ta atoms sandwiched between two layers of Se atoms in an octahedral coordination. Fig. 1b illustrates the hexagonal morphology of our 1T-$TaSe_2$ islands, indicating high epitaxial growth quality. A triangular CDW superlattice is observed on single-layer, bilayer, and trilayer 1T-$TaSe_2$, as seen in Figs. 1b, c, and

Supplementary Fig. 1 where each bright spot corresponds to a star-of-David CDW supercell. Fourier analysis of STM images (Supplementary Fig. 2) together with low-energy electron diffraction patterns (Supplementary Fig. 3) confirm the formation of a $\sqrt{13} \times \sqrt{13}$ CDW in single-layer 1T-$TaSe_2$, similar to the commensurate CDW phase of bulk 1T-$TaSe_2$ at $T < 473$ K[28] (the atomic lattice and CDW superlattice are observed to have a relative rotation angle of ~13.9°). Reflection high-energy electron diffraction patterns (Fig. 1e) and X-ray photoelectron spectroscopy (Fig. 1f) show the structural and chemical integrity of our single-layer 1T-$TaSe_2$ samples (1T and 1H islands do coexist in our samples due to the metastability of 1T-$TaSe_2$ (Supplementary Fig. 4)).

We experimentally determined the electronic structure of single-layer 1T-$TaSe_2$ in the star-of-David CDW phase using ARPES and STS. Figs. 2a and 2b show the ARPES spectra of single-layer 1T-$TaSe_2$ for *p*- and *s*-polarized light, respectively, obtained at $T = 12$ K. At low binding energies the single-layer 1T-$TaSe_2$ ARPES spectra show strongly diminished intensity at all observed momenta, indicating insulating behavior (some ARPES intensity from coexisting 1H-$TaSe_2$ islands can be seen crossing E_F at $k \approx 0.5$ Å^{-1} (white dashed lines)[29]). The CDW superlattice potential induces band folding into a smaller CDW Brillouin zone (Fig. 2b inset). One such band can be seen in the ARPES spectrum for *p*-polarized light (Fig. 2a) which shows a prominent flat band centered at $E - E_F \approx -0.26$ eV within the first CDW Brillouin zone (black dashed box). A more dispersive band can be resolved outside of the first CDW Brillouin zone boundary (vertical dashed lines labeled A and B mark this boundary). These features have no obvious photon-energy dependence (Supplementary Fig. 5) and are similar to bands observed by ARPES at the surface of bulk samples of 1T-TaS_2[16,30] and 1T-$TaSe_2$[12]. For *s*-polarized light (Fig.

2b) the flat band is much less visible and a manifold of highly dispersive bands near the Γ-point dominates the spectrum.

Our STM dI/dV spectrum (Fig. 3a (black curve)) confirms the insulating nature of single-layer 1T-$TaSe_2$. The electronic local density of states (LDOS) reflected in dI/dV exhibits a pronounced valence band peak at V = -0.33 V (labeled V_1) while dropping steeply at higher energy until reaching zero at $V \approx$ -0.05 V. The zero LDOS region bracketing the Fermi level yields an energy gap of magnitude 109 ± 18 meV (see Supplementary Fig. 6 for gap determination). The experimental LDOS does not rise again until a narrow conduction band peak is observed centered at V = 0.20 V (labeled C_1) in the empty state regime. The LDOS drops to zero again above the C_1 peak until higher-lying conduction band features are seen to rise at $V >$ 0.45 V (e.g., C_2, C_3). Aside from spatial variation in the relative peak heights, this gapped electronic structure is observed uniformly over the entire single-layer 1T-$TaSe_2$ surface (Supplementary Fig. 7). No significant band-bending effects are observed for different tip-sample separations (Supplementary Note 1 and Supplementary Figs. 8, 9). To test substrate effects we also grew single-layer 1T-$TaSe_2$ on cleaved graphite (HOPG), which shows similar STM spectra compared to single-layer 1T-$TaSe_2$/BLG (Supplementary Note 2 and Supplementary Fig. 10). This indicates that the small increase in screening provided by HOPG[21] (as shown by the slight downshift/upshift of empty-state/filled-state features in Supplementary Fig. 10) does not significantly change the 1T-$TaSe_2$ behavior. We are so far unable to experimentally test the effect of reducing screening below the level provided by BLG.

Experimental orbital texture of single-layer 1T-$TaSe_2$

To gain additional insight into the insulating ground state of single-layer 1T-$TaSe_2$, we performed dI/dV spatial mapping of its energy-dependent orbital texture at constant tip-sample

separation (Figs. 3b-h). dI/dV maps measured at negative biases all display a similar pattern where high-intensity LDOS is concentrated near the center of each star-of-David (Figs. 3b, c, and Supplementary Fig. 11). The experimental empty-state LDOS of single-layer 1T-$TaSe_2$, however, exhibits a completely different orbital texture. This is most clearly seen in the dI/dV map taken at the lowest conduction band peak C_1 (Fig. 3d) where the center of each CDW supercell is observed to be dark (i.e., no LDOS intensity). At this energy the LDOS exhibits an unusual, interlocked "flower" pattern (circled by yellow dashed lines) consisting of six well-defined "petals" (i.e., bright spots) located around the outer rim of each star-of-David. This appearance is completely different from previous reports of conduction band LDOS in bulk 1T-$TaSe_2$ and 1T-TaS_2[13,31] (which show LDOS concentrated in the star-of-David centers), and is clearly not due to defects since it follows the CDW periodicity. The 6-fold petal structure has a different symmetry than the 3-fold arrangement of top-layer Se atoms in the spaces between each star-of-David, but it shares the 6-fold symmetry of the Ta atom arrangement (Fig. 3a inset and Supplementary Fig. 12). Single-layer 1T-$TaSe_2$/HOPG shows a similar dI/dV map with the dominant LDOS intensity located near the outer rim of the stars-of-David at the lowest conduction band peak (dI/dV maps at other energies also look similar, see Supplementary Fig. 10 and Supplementary Note 2).

The dI/dV map of single-layer 1T-$TaSe_2$/BLG obtained at a slightly higher bias of V = 0.6 V (C_2) show LDOS that is related to the flower pattern in that it exhibits a nearly *inverse* flower (i.e., dark areas at C_1 are bright at C_2, see circled regions in Fig. 3e). At even higher energies the single-layer 1T-$TaSe_2$ LDOS displays other intricate orbital textures. The map at 0.8 V (Fig. 3f), for example, shows quasi-triangular patterns with intensity distributed near the outermost Ta C-atoms (labeled according to the convention shown in Fig. 1d). This evolves into trimer-like

features at 1.1 V (Fig. 3g), and a network of "rings" with intensity near Ta B-atoms at $V = 1.2$ V (Fig. 3h) (a complete set of constant-height dI/dV maps is shown in Supplementary Fig. 11).

To help quantify the complex energy-dependent LDOS distribution of single-layer 1T-$TaSe_2$, we cross-correlated our dI/dV maps with a reference map taken at the maximum of the valence band peak V_1 (Fig. 3c), which exhibits LDOS dominated by inner Ta A- and B-atoms. The resulting cross-correlation values are color-coded in Fig. 3a and show that occupied states ($-1V < V < 0$ V) all have a strong, positive cross-correlation (blue) with the valence band map at V_1 (i.e., the central Ta A- and B-atoms are bright at these energies and the C-atoms are darker). The empty-state cross-correlation, however, is very different. At C_1 (where the flower pattern is observed) the LDOS map is strongly anti-correlated (red) with the valence band map since the LDOS here is dominated by Ta C-atoms. At slightly higher energy (C_2) the cross-correlation flips to blue. This is due to the LDOS inversion that occurs at this energy (i.e., the inverse flower pattern) which creates intensity at the interior A- and B-atoms. At higher energy the cross-correlation flips again to red and stays red over a fairly wide energy range (~0.4 eV) before flipping again to blue near C_3.

Energy gap reduction and quenching of unusual orbital texture in few-layer 1T-$TaSe_2$

We examined the effect of interlayer coupling on 1T-$TaSe_2$ by studying the evolution in electronic structure as 1T-$TaSe_2$ is stacked layer by layer. We first determined the star-of-David CDW stacking order for bilayer and trilayer 1T-$TaSe_2$. As seen in the STM images of Fig. 1b and Supplementary Fig. 1, the CDW stacking order follows the shifted triclinic structure whereby inner Ta "A-atoms" sit on top of outer Ta "C-atoms", similar to stacking observed in bulk 1T-$TaSe_2$[32]. We observe that the energy gap for 1T-$TaSe_2$ narrows significantly when

interlayer coupling is added, as seen in the STM dI/dV spectra for bilayer and trilayer 1T-$TaSe_2$ shown in Fig. 4a. The bilayer energy gap reduces to 21 ± 8 meV while trilayer 1T-$TaSe_2$ shows a reduction in LDOS at E_F that can be described as "semimetallic" but exhibits no true energy gap.

In addition to reducing the 1T-$TaSe_2$ energy gap, bilayer and trilayer formation also quench the unusual orbital texture observed in the single-layer limit. As shown in the insets to Figs. 4b, c, dI/dV maps of the lowest conduction band in bilayer and trilayer 1T-$TaSe_2$ show LDOS intensity concentrated near the center of each star-of-David, in stark contrast to the flower-like orbital texture observed in single-layer 1T-$TaSe_2$ at C_1. This difference can also be seen in the color-coded cross-correlation values of bilayer and trilayer 1T-$TaSe_2$ (Figs. 4b, c). Using the valence band LDOS shown in the insets as a reference (which is similar to the single-layer valence band LDOS of Fig. 3c), the bilayer and trilayer cross-correlation remain strongly positive (blue) throughout the lowest conduction band (thus emphasizing that the LDOS here is concentrated on the interior Ta A- and B-atoms). The distinctive flower pattern seen in single-layer 1T-$TaSe_2$ at C_1 (Fig. 3d) is never seen in bilayer or trilayer LDOS at any bias (Supplementary Figs. 13, 14).

Theoretical electronic structure of single-layer 1T-$TaSe_2$ via DFT+U simulations

There are two main physical questions that we seek to answer regarding our measurements of single- and few-layer 1T-$TaSe_2$. First, what type of insulator is single-layer 1T-$TaSe_2$? And, second, what is the effect of interlayer coupling on 1T-$TaSe_2$ electronic behavior as new layers are added? To address these questions we first performed a conventional band structure calculation for freestanding single-layer 1T-$TaSe_2$ using density functional theory (DFT). From an intuitive perspective, single-layer 1T-$TaSe_2$ is expected to have metallic band structure since there are an odd number of Ta ions in the star-of-David unit cell (13) and each Ta^{4+} ion has only one *d*-electron (in principle substrate charge transfer could alter the electron counting and/or the

CDW behavior[33], but in our case charge transfer effects from the graphene substrate are negligible (Supplementary Fig. 15 and Supplementary Note 3)). As expected, the DFT band structure of single-layer 1T-$TaSe_2$ in the CDW phase calculated using the PBE exchange correlation functional shows a metallic half-filled band at E_F (Supplementary Fig. 16). This theoretical result, however, strongly disagrees with our experimental data which shows insulating behavior for single-layer 1T-$TaSe_2$ (Figs. 2, 3). An explanation for this significant discrepancy is that since the metallic band is so narrow (only ~20 meV wide) it is unstable to splitting into lower and upper Hubbard bands (LHB and UHB) due to a high on-site Coulomb energy (U) (i.e., the condition that causes Mott insulators to arise from otherwise metallic phases)[1].

To test for Mott insulator formation in single-layer 1T-$TaSe_2$ we modeled the effects of electron correlation by performing DFT+U simulations. We find that the DFT+U band structure for a ferromagnetic ground state with $U = 2$ eV reproduces most of our experimentally observed electronic structure for single-layer 1T-$TaSe_2$ (the DFT+U results were sensitive to neither the magnetic ground state nor the structural optimization conditions, and our U value is consistent with previous simulations of related systems[31,34,35] (see Supplementary Note 4 and Supplementary Figs. 17-21)). The DFT+U band structure was first compared to our ARPES data by unfolding it onto the Brillouin zone of an undistorted unit cell. As seen in Figs. 2c, d, and Supplementary Fig. 22, it reproduces the gapped electronic structure and shows good overall agreement with the ARPES spectra. In particular, DFT+U predicts that the LHB at -0.2 eV originates mainly from Ta d_{z^2} orbitals, consistent with the higher ARPES intensity under p-polarized light (Fig. 2a)[36].

The DFT+U simulations were also consistent with much of our STS data as shown in Fig. 5 which displays the simulated density-of-states spectrum and LDOS maps for single-layer 1T-$TaSe_2$. The theoretical density-of-states spectrum (Fig. 5a (black line)) reproduces the dI/dV spectrum (Fig. 3a) reasonably well in both the occupied and empty states. A LHB corresponding to the experimental V_1 feature is seen, as well as an UHB corresponding to C_1, along with higher energy features that correspond to the experimental C_2 and C_3 features. The orbital texture generated by the DFT+U calculations (Figs. 5b-h) also agree well with the experimental dI/dV maps in the valence band and upper conduction band regimes (i.e., the energies corresponding to filled states and levels above C_2).

However, there are significant discrepancies between the experimental and theoretical LDOS maps in the low-energy conduction band region ($0 < E \lesssim 0.6$ eV) where the unusual orbital texture is observed experimentally. This is most clearly seen by comparing the theoretical UHB LDOS map at 0.2 eV (Fig. 5d) with the experimental dI/dV map at $V = 0.2$ V (Fig. 3d) (i.e., the energy corresponding to C_1). The calculated LDOS has high intensity in the central Ta A- and B-atom regions (similar to what is seen in the LHB) while the experiment shows the flower pattern (i.e., the experimental LDOS occupies the Ta C-atom region and is dark in the central area). There also exists significant disagreement at the next higher energy band feature (C_2), as seen by comparison of Figs. 3e and 5e. Here the theoretical orbital texture (Fig. 5e) shows propeller-like structures with no central LDOS, while the experimental dI/dV map (Fig. 3e) shows an inverse of the C_1 flower pattern which has LDOS in the interior region of the star-of-David (a complete set of theoretical LDOS maps is shown in Supplementary Fig. 23).

Unusual empty-state orbital texture at C_1 and C_2

The good agreement between our DFT+U simulations and the majority of our ARPES and STM/STS data provides strong evidence that single-layer 1T-$TaSe_2$ is a 2D Mott insulator. However, the failure of the simulations to reproduce the unusual conduction band orbital texture at C_1/C_2 implies that additional electron-electron interactions occur in single-layer 1T-$TaSe_2$ that are not captured by DFT+U. Electrons injected from the STM tip into single-layer 1T-$TaSe_2$ at the C_1/C_2 energies experience additional correlation effects originating from their Coulomb interaction with electrons already present in the occupied electron states. Such behavior is expected to arise due to the LHB charge distribution (Fig. 3c) which creates a spatially-varying Coulomb repulsion landscape, $\tilde{U}(r)$, felt preferentially by electrons injected into UHB states since they share a common orbital. $\tilde{U}(r)$ can be estimated by treating the LHB as a Gaussian charge distribution within each star-of-David cluster and by calculating the resulting interaction energy (Supplementary Note 5). This leads to a Coulomb landscape (Supplementary Fig. 24) that is strongly repulsive to UHB electrons at the center of each star-of-David (where the LHB charge density is large) and that has minima at precisely the locations of the six-fold C_1 flower petals (where the LHB charge density is small). Given the composite nature of the UHB orbital (which has contributions from 13 Ta atoms over the CDW cell) the unusual orbital texture at C_1/C_2 can thus be understood as a redistribution of the UHB spectral density at the center of each star-of-David up to higher energy in response to enhanced Coulomb interactions that arise from reduced screening in 2D. The remaining state density of the composite UHB stays in the $\tilde{U}(r)$ minima regions and gives rise to the peripheral six-fold C_1 flower petals. This picture is corroborated by our observation that on the more strongly screened graphite substrate the LDOS distribution at C_1 of single-layer 1T-$TaSe_2$ appears to be more smeared out around the outer rim of the star-of-David cells, consistent with a less corrugated $\tilde{U}(r)$ landscape (Supplementary Fig. 10 and

Supplementary Note 2). Future theoretical treatments considering dynamical interactions could potentially provide more insight into this unusual strong correlation phenomenon.

The effect of interlayer coupling on the shifted-stacked 1T-$TaSe_2$ electronic structure is to weaken the Mott insulator phase, both in view of the observed energy gap reduction with increased layer number as well as its effect on orbital texture. The bilayer and trilayer orbital textures, for example, show no signs of the correlation-induced spectral density shift seen in the single-layer material at C_1/C_2. Such weakening of the Mott behavior likely arises from an increase in the effective inter-star-of-David hopping parameter (t) of the bilayer and trilayer due to interlayer coupling, as well as a reduction in Coulomb interactions due to increased electronic delocalization and screening.

Outlook

We have shown that single-layer 1T-$TaSe_2$ is a strongly correlated 2D Mott insulator characterized by unusual orbital texture. Interlayer coupling weakens the Mott behavior, consistent with the evolution of 1T-$TaSe_2$ into a metal as its thickness is increased layer-by-layer. The Mott insulator phase seen in single-layer 1T-$TaSe_2$ thus offers a highly-tunable 2D platform for future exploration of metal-insulator transitions[1] where the Coulomb interaction might be further modified by substrate screening[21,37], the bandwidth by pressure[38], or the carrier density by electrostatic gating[6,8].

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Author contributions

Y.C., W.R., and M.F.C. initiated and conceived the research. Y.C., W.R., H.-Z.T., R.L., S.K., F.L., and C.J. carried out STM/STS measurements and analyses. M.F.C supervised STM/STS measurements and analyses. S.T., H.R., H.X., and T.J. performed sample growth and ARPES measurements. S.-K.M., Z.-X.S., J.A.S., and Z.L. supervised sample growth and ARPES measurements. M.W. performed DFT calculations and theoretical analyses. S.G.L. supervised DFT calculations and theoretical analyses. J.E.M. performed electrostatic modeling. O.R.A. and A.L. provided support for development of the CDW model. Y.C., W.R., and M.F.C. wrote the manuscript with the help from all authors. All authors contributed to the scientific discussion.

Competing interests

The authors declare no competing interests.

Data availability

The data represented in Figs. 1f, 3a, 4, and 5a are available as source data in Supplementary Data 1-4. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

Code availability

The codes used in this study are available from the corresponding author upon reasonable request.

Methods

Sample growth and ARPES measurements

Single-layer 1T-$TaSe_2$ films were grown on epitaxial bilayer graphene terminated 6H-SiC(0001) and cleaved HOPG substrates in a molecular beam epitaxy chamber operating at ultrahigh vacuum (UHV, base pressure 2×10^{-10} Torr) at the HERS endstation of Beamline 10.0.1, Advanced Light Source, Lawrence Berkeley National Laboratory. High purity Ta (99.9%) and Se (99.999%) were evaporated from an electron-beam evaporator and a standard Knudsen cell, respectively, with a Ta:Se flux ratio set between 1:10 and 1:20 and a substrate temperature of 660 $^{\circ}$C. A higher substrate temperature (compared with our previous 1H-$TaSe_2$ growth at 550 $^{\circ}$C[29]) was used to facilitate the growth of the metastable 1T phase of $TaSe_2$. The growth process was monitored by reflection high-energy electron diffraction. After growth, the films were

transferred *in-situ* into the analysis chamber (base pressure 3×10^{-11} Torr) for ARPES and core-level spectra measurements. The ARPES system was equipped with a Scienta R4000 electron analyzer. The photon energy was set at 51 eV (unless specified otherwise) with energy and angular resolution of 12 meV and 0.1°, respectively. *p*- and *s*-polarized light were used, as described elsewhere (ref. [39]). Before taking the films out of vacuum for STM/STS measurements, Se capping layers with ~10 nm thickness were deposited onto the samples for protection. These were later removed by UHV annealing at ~200 °C for 3 hours.

STM/STS measurements

STM/STS measurements were performed using a commercial CreaTec STM/AFM system at T = 5 K under UHV conditions. To avoid tip artifacts, STM tips were calibrated on a Au(111) surface by measuring its herringbone surface reconstruction and Shockley surface state before all STM/STS measurements. Both W and Pt-Ir STM tips were used and yielded similar results. STS dI/dV spectra were obtained using standard lock-in techniques with a small bias modulation at 401 Hz. The constant-height mode (i.e., feedback loop open) was used for collecting all dI/dV conductance maps. Before obtaining each set of maps the STM tip was parked near the sample surface for at least 8 hours to minimize piezoelectric drift effects.

Electronic structure calculations

First-principles calculations of single-layer 1T-$TaSe_2$ were performed using density functional theory (DFT) as implemented in the Quantum ESPRESSO package[40]. The onsite Hubbard interaction was added through the simplified rotationally invariant approach using the same U value for each Ta atom[41,42]. A slab model with 16 Å vacuum layer was adopted to avoid interactions between periodic images. We employed optimized norm-conserving Vanderbilt pseudopotentials (ONCVPSP) including Ta 5*s* and 5*p* semicore states (with a plane-wave energy

cutoff of 90 Ry)[43-45] as well as the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional[46] in the generalized gradient approximation (GGA). The structure was fully relaxed at the DFT-PBE level until the force on each atom was less than 0.02 eV/Å (unless specified otherwise). The resulting relaxed single-layer 1T-$TaSe_2$ in the $\sqrt{13} \times \sqrt{13}$ CDW phase has a lattice constant of a = 12.63 Å. Spin-orbit coupling was not taken into account in our calculations since it has a negligible influence on the band structure given the inversion symmetry of this system. The unfolding of the band structure from the CDW supercell to the undistorted unit cell was calculated using the BandUP code[47,48] with band energies and wavefunctions obtained from the Quantum ESPRESSO package.

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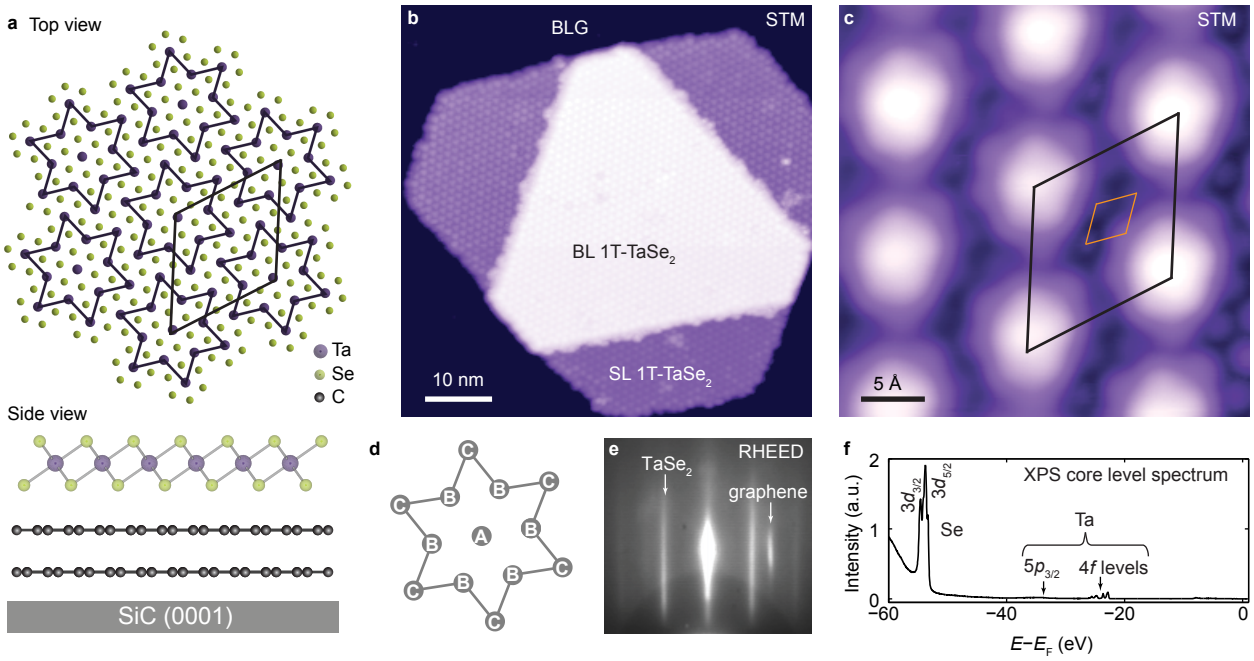
Fig. 1. Structure of single-layer 1T-$TaSe_2$ in the star-of-David CDW phase. a, Top and side view sketches of single-layer 1T-$TaSe_2$, including substrate. Clusters of 13 Ta atoms in star-of-David CDW supercells are outlined, as well as the CDW unit cell. **b**, Large-scale STM topograph of a typical 1T-$TaSe_2$ island shows monolayer and bilayer regions (V_b = -0.5 V, I_t = 10 pA, T = 5 K). **c**, A close-up STM image of single-layer 1T-$TaSe_2$. Each bright spot corresponds to a star-of-David supercell (V_b = -0.17 V, I_t = 3 nA, T = 5 K). Black and orange parallelograms mark CDW and atomic unit cells, respectively. **d**, Labels for Ta atoms in the star-of-David CDW supercell depend on radial distance from center. **e**, Reflection high-energy electron diffraction pattern of a submonolayer 1T-$TaSe_2$ film. **f**, X-ray photoelectron spectroscopy shows characteristic peaks of Ta and Se core levels for a submonolayer 1T-$TaSe_2$ film.

Fig. 2. ARPES and DFT+U band structure of single-layer 1T-$TaSe_2$. ARPES spectra of single-layer 1T-$TaSe_2$ acquired with **a,** *p*- and **b,** *s*-polarized light at $T = 12$ K along the Γ-K' and Γ-M' directions defined in the undistorted (i.e., no CDW) unit cell Brillouin zone (yellow hexagon in Fig. 2b inset). ARPES spectra have little intensity at low binding energies except for coexisting 1H-$TaSe_2$ bands that cross E_F at $k \approx 0.5$ Å^{-1} (white dashed lines). A strong flat band is seen under *p*-polarized light in the first CDW Brillouin zone (black dashed box in **a**). The full CDW Brillouin zone is sketched in the inset of **b** (black hexagon). **c**, DFT+U band structure ($U =$ 2 eV) of single-layer 1T-$TaSe_2$ unfolded onto the undistorted unit cell Brillouin zone compared to ARPES spectrum under *p*-polarized light (from **a**). **d**, Same DFT+U band structure as in **c** compared to ARPES spectrum under *s*-polarized light (from **b**).

Fig. 3. Experimental energy-resolved unusual orbital texture of single-layer 1T-$TaSe_2$. a, STS dI/dV spectrum of single-layer 1T-$TaSe_2$ shows a full energy gap bracketed by two STS peaks labeled V_1 and C_1 (f = 401 Hz, I_t = 50 pA, V_{RMS} = 20 mV). Color shows cross-correlation of dI/dV maps at different energies with the reference map shown in **c**. Inset shows how the unusual orbital texture in **d** compares to atomic site locations (the 6-fold petal structure is shaded gray in the inset). **b**-**h**, Constant-height dI/dV conductance maps of the same area for different bias voltages show energy-dependent orbital texture (f = 401 Hz, V_{RMS} = 20 mV). The same star-of-David CDW supercell is outlined in each map (orange line). Yellow dashed circles in **d**, **e** highlight the unusual LDOS patterns at C_1 and C_2 and their relative spatial inversion.

Fig. 4. Energy gap reduction and quenching of unusual orbital texture in few-layer 1T-$TaSe_2$. a, STS dI/dV spectra for single-layer, bilayer, and trilayer 1T-$TaSe_2$ show how interlayer coupling reduces the energy gap with an increasing number of layers. Spectra are shifted vertically for viewing (horizontal dashed lines mark dI/dV = 0, f = 401 Hz, V_{RMS} = 2 mV). dI/dV maps of the valence and conduction band LDOS as well as larger energy-scale dI/dV spectra of **b,** bilayer, **c,** trilayer 1T-$TaSe_2$ (f = 401 Hz, V_{RMS} = 20 mV). Spatial cross-correlation values are shown color-coded with references taken near the LDOS maximum of the valence band for bilayer and trilayer 1T-$TaSe_2$. In contrast to single-layer 1T-$TaSe_2$, the lowest conduction band for both bilayer or trilayer show no unusual orbital texture, thus resulting in positive cross-correlation values (blue), indicating that LDOS is concentrated on the interior Ta A- and B-atoms.

Fig. 5. Theoretical orbital texture of single-layer 1T-$TaSe_2$ from DFT+U simulations. a, Theoretical density of states of single-layer 1T-$TaSe_2$ from DFT+U simulations (U = 2 eV). Color shows cross-correlation of LDOS maps at different energies with respect to the reference map in **c** (-0.2 eV). **b**-**h**, Theoretical LDOS maps of single-layer 1T-$TaSe_2$ from DFT+U simulations (U = 2 eV). The same star-of-David supercell is outlined in each map (orange line). Yellow dashed circles in **d**, **e** highlight two star-of-David clusters which show very different theoretical conduction band orbital texture compared to experimental C_1 and C_2 features in Figs. 3d, e.


a Top view
Ta
Se
C
Side view
SiC (0001)
b
BLG
STM
BL 1T-TaSe2
10 nm
SL 1T-TaSe2
c
STM
5 Å
d
A
B
C
e
TaSe2
RHEED
graphene
f
XPS core level spectrum
3d3/2
3d5/2
Se
Ta
5p3/2
4f levels
Intensity (a.u.)
2
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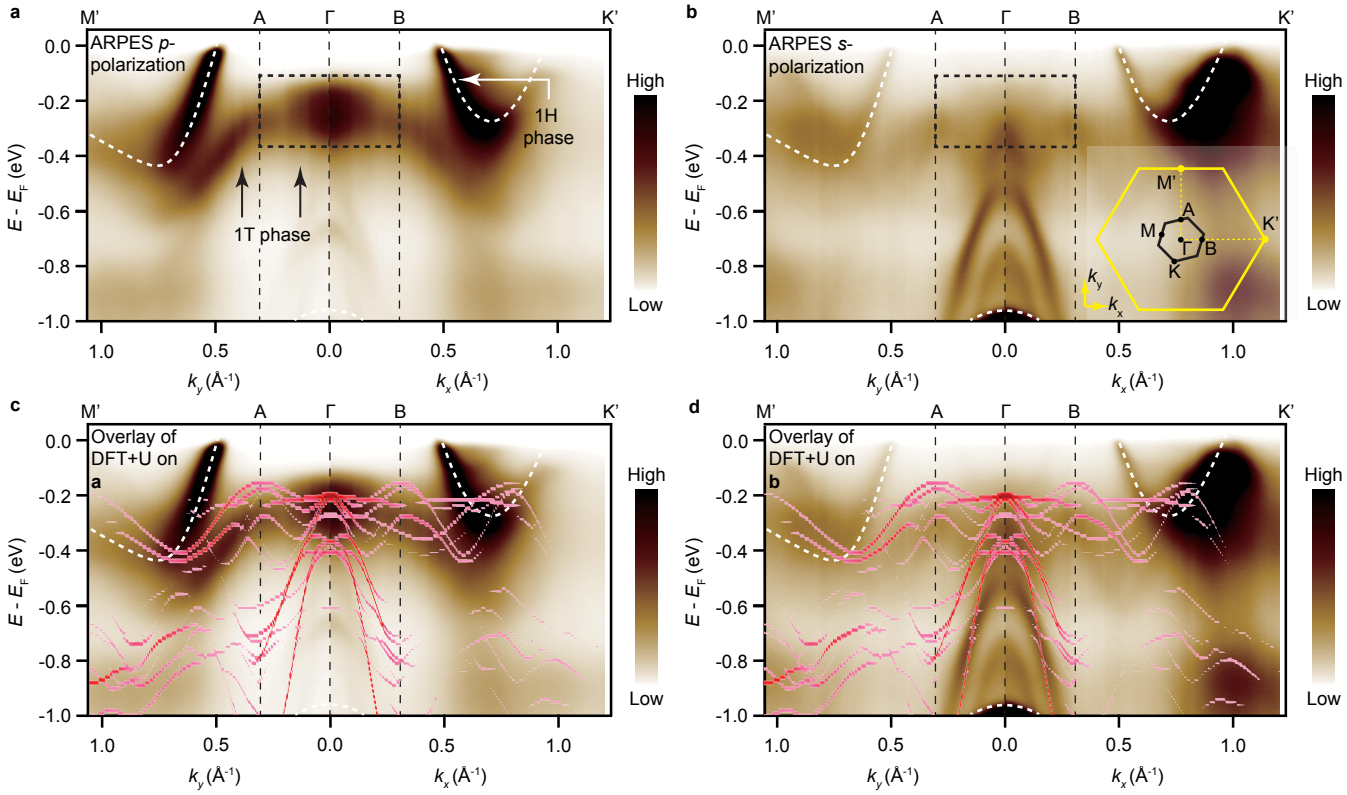
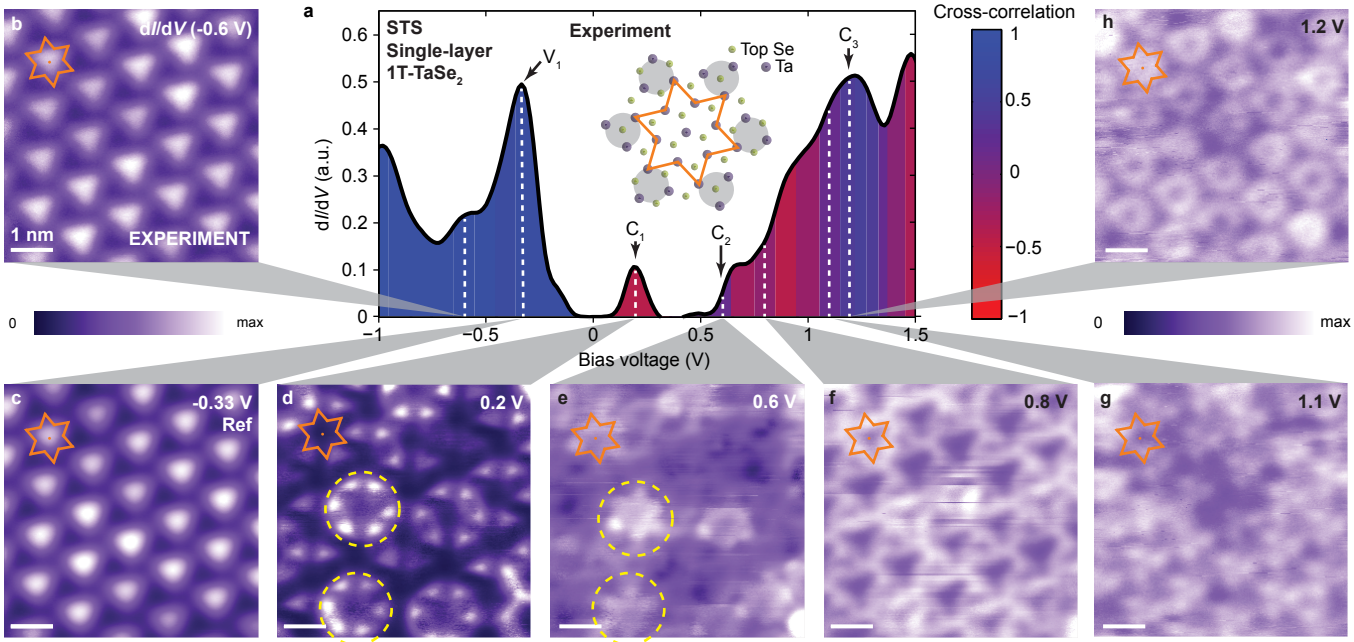
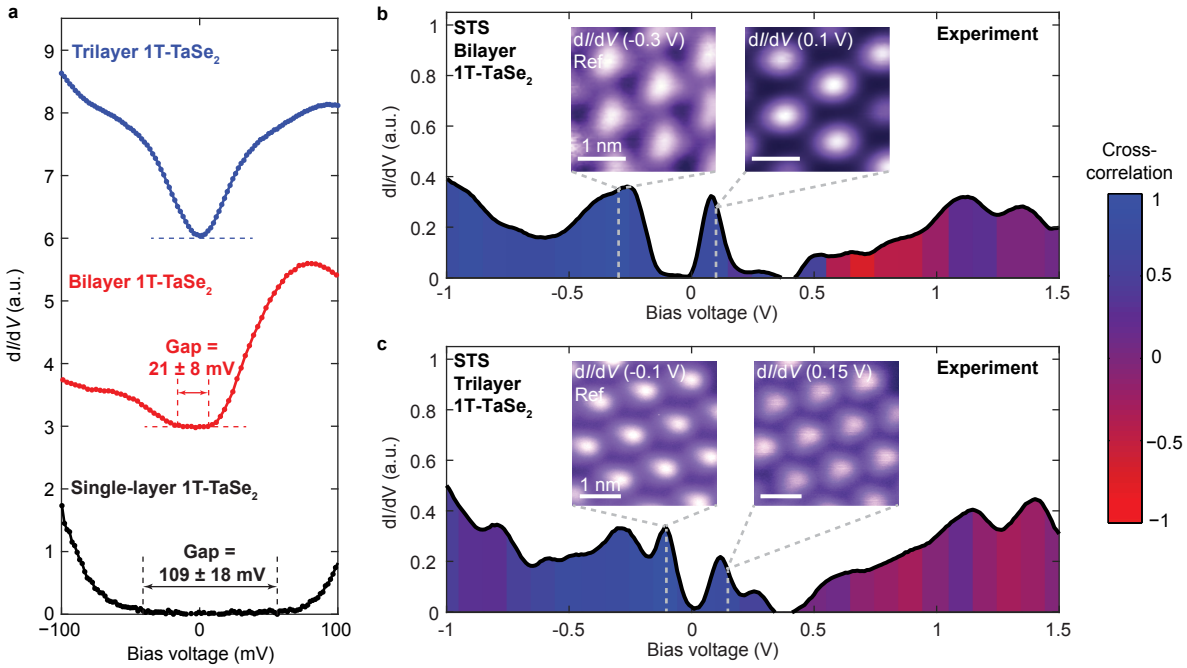
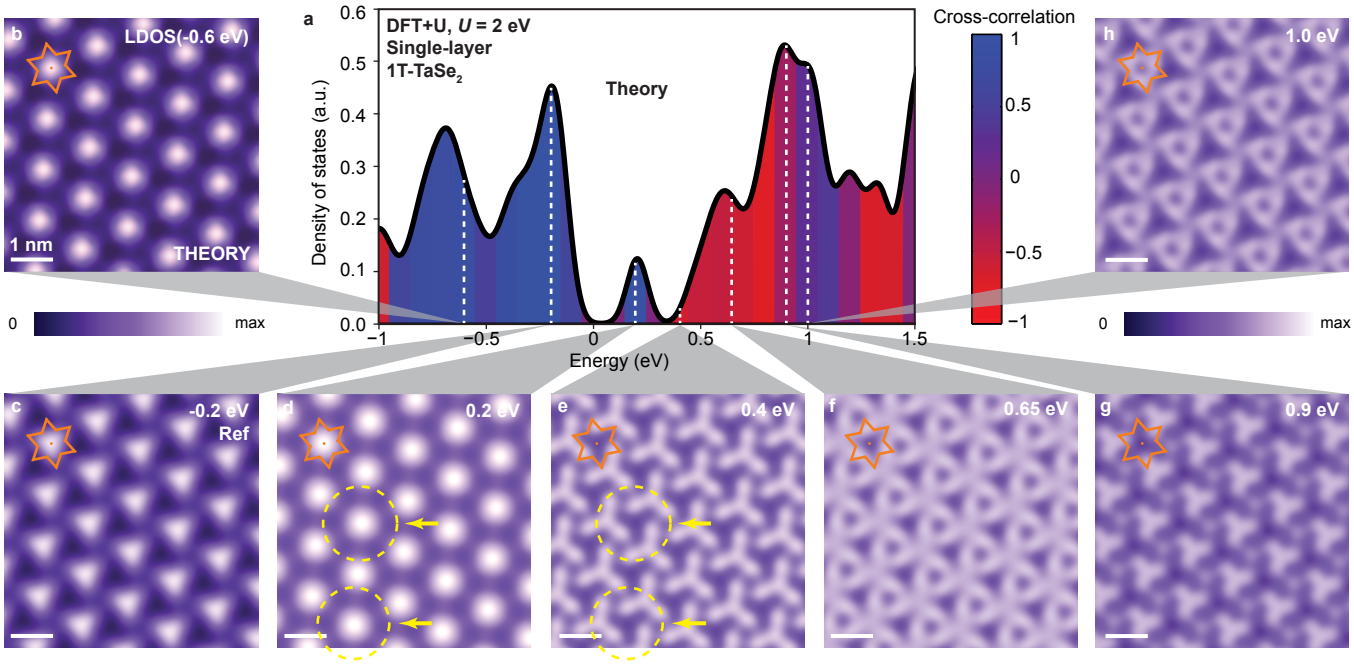
0
−60
−40
−20
0
E−EF (eV)


a
M'
A
Γ
B
K'
ARPES p-polarization
1H phase
1T phase
High
Low
E - E_F (eV)
k_y (Å^-1)
k_x (Å^-1)
b
ARPES s-polarization
c
Overlay of DFT+U on a
d
Overlay of DFT+U on b


b
dI/dV (-0.6 V)
1 nm
EXPERIMENT
0
max
a
STS
Single-layer
1T-TaSe2
Experiment
Top Se
Ta
V1
C1
C2
C3
dI/dV (a.u.)
Bias voltage (V)
Cross-correlation
1
0.5
0
−0.5
−1
h
1.2 V
0
max
c
-0.33 V
Ref
d
0.2 V
e
0.6 V
f
0.8 V
g
1.1 V


a
Trilayer 1T-TaSe2
Bilayer 1T-TaSe2
Gap =
21 ± 8 mV
Single-layer 1T-TaSe2
Gap =
109 ± 18 mV
dI/dV (a.u.)
Bias voltage (mV)
b
STS
Bilayer
1T-TaSe2
dI/dV (-0.3 V)
Ref.
1 nm
dI/dV (0.1 V)
Experiment
dI/dV (a.u.)
Bias voltage (V)
c
STS
Trilayer
1T-TaSe2
dI/dV (-0.1 V)
Ref.
1 nm
dI/dV (0.15 V)
Experiment
dI/dV (a.u.)
Bias voltage (V)
Cross-
correlation


a
DFT+U, U = 2 eV
Single-layer
1T-TaSe2
Theory
Density of states (a.u.)
Energy (eV)
Cross-correlation
b
LDOS(-0.6 eV)
1 nm
THEORY
0
max
h
1.0 eV
c
-0.2 eV
Ref
d
0.2 eV
e
0.4 eV
f
0.65 eV
g
0.9 eV